Superconducting fluctuations in small grains - the Universal Hamiltonian and the reduced BCS model

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Abstract

Small superconducting grains are discussed in the frameworks of both the reduced BCS Hamiltonian and the Universal Hamiltonian. It is shown that fluctuations of electrons in levels far from the Fermi energy dominate superconducting properties in small and ultrasmall grains. Experimental consequences related to the spin susceptibility and persistent currents of grains and rings with weak electron-electron interactions are discussed.

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1 Introduction

The paradigmatic framework in the study of superconducting fluctuations in small grains[1] is the reduced BCS Hamiltonian

$$\hat{H} = \sum_{j,\sigma=\pm} \epsilon_j c^\dagger_{j\sigma} c_{j\sigma} - \lambda d \sum_{i,j} c^\dagger_{i+} c^\dagger_{i-} c_{j-} c_{j+}. \tag{1}$$

Here λ is the dimensionless interaction constant, d is the mean level spacing, and the indices i, j correspond to doubly degenerate time reversed states of energy $E_{\mathrm{F}} - \omega_{\mathrm{D}} < \epsilon < E_{\mathrm{F}} + \omega_{\mathrm{D}}$.

While the mean field (BCS)[2] solution of this Hamiltonian was extremely powerful in explaining the properties of bulk superconductors, grains of finite size pose additional challenges. In such grains the number of electrons is fixed, their size may be smaller than the coherence length ξ and the magnetic

penetration depth, and zero resistance is not achievable. Still, already in 1959 Anderson postulated that superconducting grains maintain superconducting properties for sizes much smaller than ξ, down to the size in which the single electron level spacing d equals the bulk energy gap Δ (the "Anderson size"[3]). For clean grains this size is of order $\xi^{1/3}\lambda_{\mathrm{F}}^{2/3} \ll \xi$.

Much of the recent theoretical interest in the study of small superconducting grains (see Ref.[1] and references therein) was initiated by the experimental work of Ralph Black and Tinkham[4], who found a parity dependent gap in the excitation spectrum of single "small" Al grains (with $d < \Delta$), but not for "ultrasmall" grains having $d > \Delta$. More recently it was also shown that the Meissner effect sharply disappears at grain sizes consistent with the Anderson size[5,6,7].

While superconducting characteristics reminiscent of bulk properties indeed vanish at the Anderson size, signatures of pairing correlations persist to smaller sizes. These signatures are a result of superconducting fluctuations of electrons at energies larger than the gap energy, up to the cutoff energy given by ω_{D}. For example, the spin susceptibility of ultrasmall superconducting grains is predicted to exhibit a re-entrant behavior both as function of temperature[8] and as function of magnetic field[9,10], with a long tail, persisting up to the temperature/magnetic field equivalent of ω_{D}. The superconducting fluctuations of electrons further than Δ from the Fermi energy E_{F} affect significantly also grains in an intermediate regime, where $\Delta^2/\omega_{\mathrm{D}} < d < \Delta$. In particular, in this regime the energy gain of the system by the attractive interaction is much larger than that given by the mean field treatment of BCS[9], and the same is true for the pairing parameter[11]. Moreover, it was shown that these far level fluctuations affect differently single particle and collective properties, and therefore one has to define correspondingly two different pairing parameters[11] when discussing small superconducting grains.

While the above properties, and specifically their dependence on the high energy cutoff, stem from the exact solution of the reduced BCS hamiltonian[12,13], the question to their relevance to real superconducting grains may arise[14]. This question is of particular relevance since specific predictions for the experimental consequences of the fluctuations of the far levels (e.g. the long tail of the spin susceptibility discussed above) are made. Here we discuss the significance of the superconducting fluctuations of the far levels by considering both the reduced BCS model and the model of the Universal hamiltonian[15]. While the reduced BCS Hamiltonian is an effective Hamiltonian whose validity in describing high energy properties may be questioned, the Universal Hamiltonian was shown, using renormalization group approach, to control the low energy physics of metallic grains with weak interactions and large dimensionless conductance[16,17]. We thus establish the significance of the superconducting fluctuations of the far levels up to the high energy cutoff of the

Universal Hamiltonian given by the Thouless energy $E_{\rm Th}$. We then discuss the significance of the Debye energy in general, and for the problem of persistent currents in particular.

2 Reduced BCS Hamiltonian and Universal Hamiltonian - exact solution

The reduced BCS Hamiltonian (1), with finite number of electrons, was solved exactly by Richardson[12,13]. The structure of the solution takes advantage of the fact that the interaction scatters only pairs and not single electrons. Thus, the solution is obtained separately for subspaces of the Hilbert space defined by the identity of the singly occupied levels. The singly occupied levels are neglected at first, the problem of N pairs in M states is solved, and the singly occupied levels are then trivially added to the solution (see Refs.[12,13] as well as Ref.[18] for details).

While the reduced BCS Hamiltonian is motivated by the specific phonon-mediated attractive electron-electron interaction, it was recently shown that any metallic grains with large dimensionless conductance $g \equiv E_{\rm Th}/d$, weak interactions, and negligible spin-orbit interaction, can be described by the Universal Hamiltonian[15], which includes only three interaction parameters

$$H = \sum_{n,\sigma} \epsilon_n c^\dagger_{n,\sigma} c_{n,\sigma} + E_c \hat{N}^2 + J_c \hat{T}^\dagger \hat{T} + J_s \hat{S}^2. \tag{2}$$

Here $\hat{N} = \sum_{n,\sigma} c^\dagger_{n,\sigma} c_{n,\sigma}$ is the number operator, $\hat{\vec{S}} = \frac{1}{2}\sum_{n,\sigma,\sigma'} c^\dagger_{n,\sigma} \vec{\sigma}_{\sigma,\sigma'} c_{n,\sigma'}$ is the total spin operator, and $\hat{T} = \sum_n c_{n,-} c_{n,+}$ is the pair annihilation operator. The index n spans a shell of doubly degenerate time reversed states of energy $E_{\rm F} - E_{\rm Th} < \epsilon_n < E_{\rm F} + E_{\rm Th}$. E_c is the charging energy and $J_{c(s)} = \lambda_{c(s)} d$, where λ_c and λ_s are the dimensionless interaction parameters in the Cooper channel and in the spin channel respectively.

Interestingly, for isolated grains the solution of the Universal Hamiltonian is given by Richardson's solution for the reduced BCS Hamiltonian. First, the Coulomb term can be neglected, since the number of electrons in the isolated grain remains constant. Then, other than a different cutoff energy ($E_{\rm Th}$ compared to $\omega_{\rm D}$), one remains with the reduced BCS Hamiltonian with the additional exchange term. The latter commutes with the rest of the Hamiltonian. Moreover, the pairing interaction involves solely the paired levels, while the spin interaction involves solely the singly occupied levels. One can then obtain the solution for the Universal Hamiltonian by following the steps in Richardson's solution of the reduced BCS Hamiltonian, with the additional

consideration of the spin term for the singly occupied levels.

3 Large contribution of the far levels

Considering the reduced BCS Hamiltonian (1), and using Richardson's exact solution, the condensation energy of small metallic grains was calculated as function of the coupling constant λ[9]. The condensation energy was defined as the difference between the energy of the Fermi state and the real ground state of the system, i.e.

$$E_{\mathrm{cond}}(\lambda) \equiv E_{\mathrm{F.g.s}}(\lambda) - E_{\mathrm{g.s.}}(\lambda). \tag{3}$$

For bulk superconductors this energy is given by $\Delta^2/(2d)$, is extensive, and is a non-analytic function of λ. For finite size grains it was found that the condensation energy is analytic at $\lambda = 0$, and is very well estimated by $E_{\mathrm{cond}} \simeq \Delta^2/(2d) + \ln 2\lambda^2\omega_{\mathrm{D}}$. The first term gives the contribution of the levels within Δ of E_{F}, and the second, perturbative term, is the contribution of the far levels. Interestingly, due to unique analytical properties of the condensation energy, the perturbative term expresses the contribution of the far levels not only within the regime of validity of perturbation theory ($\lambda < 1/\ln N$, or $d > \Delta$), but also in the regime where $d < \Delta$[9]. An immediate outcome of this result is that for a large intermediate regime, $\Delta^2/\omega_{\mathrm{D}} < d < \Delta$, the condensation energy is much larger than that given by the BCS term. Note that the perturbative term is intensive, and therefore negligible as the size of the grain becomes large.

Similarly, the contribution of the far levels is found to be significant when considering generalizations of the bulk order parameter that are suitable for finite size grains[11] (see also Ref.[1] and references therein). For all standard definitions of the order parameter the far level contribution results in a term linear in ω_{D}, and the order parameter being much larger than its mean field value in the intermediate regime defined above (see Ref.[11] for details). Thus, in contrast to the common belief that the order parameter turns from being extensive to being intensive at the Anderson size ($d \approx \Delta$), it actually becomes intensive already at a much larger size, i.e. when $d \approx \Delta^2/\omega_{\mathrm{D}}$. Furthermore, the significant contribution of the far levels separates collective properties of the superconductor such as the condensation energy, from single particle properties such as the energy gap for single particle excitations. While in bulk superconductors both energies are related to the bulk order parameter Δ, for small grains one has to define two different order parameters to describe collective and single particle properties[11]. While the cutoff energy affects the former linearly, it affects single particle properties only logarithmically.

Interestingly, using the similarity of the reduced BCS and the Universal Hamiltonians, and their exact solution, the results discussed above for the reduced BCS Hamiltonain are immediately applicable to the Universal Hamiltonian, with the only change of the cutoff from $\omega_{\rm D}$ to $E_{\rm Th}$. Thus, as long as the grains obey the conditions of applicability of the Universal Hamiltonian described above, one obtains the intermediate regime, defined now by $\Delta^2/E_{\rm Th} < d < \Delta$, in which e.g. the condensation energy is much larger than the mean field BCS value, and therefore intrinsic. The predictions of re-entrant spin susceptibility as function of temperature[8] and magnetic field[9,10], which result from the pairing correlations of the far level, can also be made on the basis of the validity of the Universal Hamiltonian. Importantly, all the above characteristics do not depend on the interaction being constant up to the upper cutoff, nor do they depend on the existence of a sharp cutoff. It is sufficient that one can bound the interaction from below by $c_1\lambda_c$ within a window of $c_2E_{\rm Th}$ from $E_{\rm F}$, where c_1, c_2 are constants of order unity. Thus, based on the validity of the Universal Hamiltonian one can argue for the applicability of the above results for the condensation energy and spin susceptibility for real physical systems. Note, that within the regime of applicability of the Universal Hamiltonian one can uniquely relate the excess spin susceptibility as function of the magnetic field to the existence of pairing correlations, as the exchange term can not account for such behavior. Furthermore, by examining the behavior of the spin susceptibility of an ensemble of small metallic grains the interaction parameters of the Universal Hamiltonian can be determined[10]. Another consequence of the above analysis is that the interesting question of whether the noble metals have a weak attractive interaction and therefore are superconductors albeit with a very low T_c can be addressed as well, in small grains and by extrapolation in bulk. This is since unlike T_c which is exponentially small in λ_c, the excess spin susceptibility as a function of the magnetic field is quadratic in λ_c and therefore detectable for small λ_c.

4 Persistent currents and the significance of $\omega_{\rm D}$

In the discussion above it is argued that the interesting physics related to the pairing correlations of the far levels is independent from the validity of the reduced BCS model in describing the properties of small grains at energies of the order of $\omega_{\rm D}$. Still, the energy scale of $\omega_{\rm D}$ is a physical energy scale, related to the retardation of the phonon mediated interaction. Especially since, as was mentioned above for the Universal Hamiltonian, a constant interaction and a sharp cutoff at $\omega_{\rm D}$ are not necessary for the applicability of the above results, it is plausible that indeed the energy scale of $\omega_{\rm D}$ dictates the magnitude of the condensation energy of small grains, the boundary of the intermediate regime, and the extent of the tail in the excess spin susceptibility.

However, in relation to persistent currents, a rigorous understanding of the interaction is even more crucial. Since the persistent current in a normal metal ring is given by the derivative of the energy with respect to the flux, it is plausible that for metals with weak attractive interaction, the large contribution of the far levels to the condensation energy will affect the persistent current, and will result, within the reduced BCS model, in a large persistent current related to the energy scale of $\omega_{\rm D}$. Indeed, in Ref. [19] it was shown that the value of the derivative of the persistent current at zero flux is much larger within the reduced BCS model in comparison to the value obtained within the standard theory of momentum independent interaction[20]. Here, however, the difference in magnitude of the persistent current is related to the high energy cutoff being $\omega_{\rm D}$ rather than $E_{\rm Th}$, and thus is crucially related to the specifics of the model considered, and to the exact form of the interaction. In particular, the BCS interaction assumes that only time reversed states interact, i.e. that the total incoming momentum of the scattered electrons q must be zero. While this is a simplified form of the momentum dependence of the interaction, the subsequent result of the much larger magnetic reponse[19] in comparison to the value obtained within the momentum independent picture points to the importance of taking correctly the dependence of the interaction on q, especially since a significant q dependence of the attractive interaction is motivated by the retardation of the phonon mediated interaction[19,21]. We believe that a rigorous understanding of the form of the interaction, and in particular its q dependence, could lead to a better understanding of the long standing question regarding the value of the ensemble averaged persistent current in small metallic rings.

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